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Effects of One-Dimensional Stress on MIL-STD-1376 Piezoelectric Ceramic Materials, Types I, II, and III

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The effects of one-dimensional compressive stress parallel to the polarized axis of ferroelectric ceramics Type I, II, and III (MIL-STD-1376) were tested. Material of each type was obtained from five manufacturers and tested to determine the dependence on stress of the constants g_{33}, d_{33}, and K_{33}^{T} and dielectric loss tangent. Data and relative comparisons between material types were determined.

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CONTENTS

EFFECTS OF ONE-DIMENSIONAL STRESS ON MIL-STD-1376 PIEZOELECTRIC CERAMIC MATERIALS, TYPES I, II, AND III

INTRODUCTION

It is important to know the effects of compressive stress on polarized ferroelectric ceramics because their characteristics tend to depend largely on past history, and particularly on temperature and stress. These ceramic materials are used by the U.S. Navy in most underwater sound transducers for reasons that include good piezoelectric activity, high dielectric constant, mechanical adaptability, and relatively low cost. In a practical transducer design, the stress may be one-, two-, or three-dimensional and parallel or normal to the axis of polarization. This report is concerned with the effects of one-dimensional compressive stress parallel to the polarized axis on commercial materials of MIL-STD-1376, Types I, II, and III [1].

Ceramics were purchased from Channel Industries, Edo Western, Gulton Industries, Marine Resources, Inc., Vernitron Piezoelectric Division, and Honeywell Ceramics under their trade name designations which should be equivalent to Types I, II, and III. These ceramics were tested to determine the effects of stress on parameters K_{33}^T, tan δ, g_{33}, and d_{33}. Although the data and conclusions of this report apply only to the samples on which measurements were made, the results of the samples of the same type from different manufacturers fell into ranges that sometimes overlapped those of other types.

In previous work [2], the amount of variation in the data due to manufacturer's batch variation was unknown. The present work provides data on ceramics purchased from Gulton and Vernitron 2 years later, the results for which showed fair agreement with the previous work. Thus, a preliminary indication is that batch variation of data on the reaction to stress of ceramics from a specific manufacturer does not appear to be great.

BACKGROUND

The mechanical, dielectric, and piezoelectric properties of piezoelectric ceramics vary because of different compositions, additives, and production procedures. MIL-STD-1376 was written in 1970 to classify the various ceramics into four general groupings according to ranges of certain material properties. Types I, II, and III are lead zirconate titanate (PZT) ceramics with a nominal zirconate/titanate ratio of 53/47 (modified with additives). The types differ according to Curie point and additives; the Curie point is above 310°C for Type I, above 330°C for Type II, and above 290°C for Type III. Each type has additives consistent with its special material properties. Type IV ceramics are of barium titanate with special additives.

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MIL-STD-1376 covers the properties, quality requirements, and testing of piezoelectric ceramics for use in Navy Sonar Transducers, including hydrophones. The small signal properties to be measured using the ceramic standard test specimen include dielectric constant K_{33}^T, dielectric loss tangent tan δ, piezoelectric coupling factor (effective) k_{eff}, frequency constant N_1, density ρ, mechanical quality factor Q_m, and temperature change of K_{33}^T. Tests to determine the aging rate of certain properties are conducted in the period 10 to 100 days after poling. The effects of stress on the ceramics are not specified, however.

The effects of compressional stress on piezoelectric ceramics have been described by several investigators [2-13]. Most of these studies have been limited to a maximum stress of 138 MPa (20 000 psi). This report is a continuation of the work by Meeks and Timme [2,12,13] to completely characterize the effects of stress amplitude, orientation, and dimensionality on Types I, II, and III materials. The approach is to understand how stresses individually and collectively act to affect the characteristics of the ceramic.

MEASUREMENT METHOD

Measurements of piezoelectric constants g_{33} and d_{33}, relative dielectric constant K_{33}^T, and tan δ were made as a function of stress in a reciprocity-coupler chamber [14] using the technique developed by Meeks and Timme [13]. The description of the electronics system and method of calculating the results is reported separately [15]. The piezoelectric constant is given by the expression

$$g_{33} = \frac{1}{4t}\left(\frac{2\pi f V R e_{CA} e_{CB}}{\rho c^2 e_A e_{AB}}\right)^{1/2} \tag{1}$$

where t is the ceramic thickness; f is the frequency; V is the reciprocity-coupler volume; R is the current resistor value; ρ is the castor oil density; c is the speed of sound in castor oil; and e_{CA}, e_{CB}, e_A, and e_{AB} are four voltages involved in the reciprocity measurement. The factor of 4 in the denominator is a surface area ratio that comes from the method of mounting the ceramic sample in a test fixture. The relative dielectric constant K_{33}^T is related to the capacitance by the expression

$$K_{33}^T = Ct/A\epsilon_0 \tag{2}$$

where ϵ_0 is the permittivity of free space, C is the capacitance, t is the thickness, and A is the cross-sectional area of the ceramic specimen. Dissipation factor tan δ and capacitance C are measured with a capacitance bridge using the three-terminal method. Piezoelectric constant d_{33} is calculated using the expression

$$d_{33} = g_{33} K_{33}^T \epsilon_0 . \tag{3}$$

The specimen holder and method of preparing the ceramic sample are as reported by Meeks and Timme [13]. The maximum hydrostatic pressure in the test chamber was about 80 MPa, but the end cap design of the specimen holder increased the magnitude of the stress on the ceramic sample by a factor of 4 and converted it from hydrostatic to single dimensional.

RESULTS AND DIMENSIONS

Test results comparing ceramics of the same type emphasize similarities. The figures showing representative curves for g_{33}, d_{33}, K_{33}^T, and tan δ for the various samples include data through the first full stress cycle; the recovery on release of stress is an indication of the resistance of the ceramic to depolarization. Data for g_{33}, d_{33}, and K_{33}^T have been normalized to initial values and plotted against the log of stress T_3. The normalized K_{33}^T and tan δ are plotted linearly against log T_3, while normalized g_{33} and d_{33} are further reduced to a dB [20 log (normalized value)] presentation because this is the way that transducer designers generally use the information.

Certain general similarities appear in the data. Dielectric constant K_{33}^T always increases to a maximum with increasing stress, and thereafter decreases approximately as the inverse cube root of the stress. The stress at which K_{33}^T becomes maximum differs from type to type and to a lesser extent within each type. Dissipation factor tan δ is somewhat different for each of the three ceramic types, but similar within each type. Piezoelectric constant g_{33} is relatively independent of applied stress up to a point that is different for each of the three ceramic types. Above this stress the g_{33} constant decreases approximately as the inverse 5/3 power of stress. The piezoelectric constant d_{33}, which is calculated from Eq. (3) and thus contains the stress dependencies of both K_{33}^T and g_{33}, increases to a maximum with applied stress and then decreases as the inverse square of the stress.

Type I Ceramics

Table 1 reviews the five Type I materials studied. Dielectric constant K_{33}^T peaked at stresses between 93 and 140 MPa, with an average of 114 MPa. The percentage change of K_{33}^T from initial value to peak ranged from +54% to +95%, with an average of 65%. The dissipation factor tan δ peaked on most samples around 100 MPa with a value less than 0.009.

Table 1 — Comparison of Type I Ceramics

Manufacturer	Mfg. Type	Stress K_{33}^T (max) (MPa)	K_{33}^T Percentage Change Initial-To-Peak	tan δ		Stress g_{33} (-3 dB) (MPa)	Stress d_{33} (-3 dB) (MPa)
				Max.	Min.		
Channel	5400	93	+55	0.0079	0.0013	95	132
Gulton	HDT-31	140	+54	0.0084	0.0050	130	178
Marine Resources	TCD-4	102	+56	0.0088	0.0048	103	135
Vernitron	PZT-4	132	+65	0.0061	0.0027	125	165
Honeywell	K-12	105	+95	0.0066	0.0012	93	155
Average		114	65			109	153

For g_{33} and d_{33} the stress value indicated is the point at which the parameter value decreased to 0.707 of the initial value; or this corresponds to a decrease of 3 dB for receiving sensitivity in a hydrophone application. For g_{33} the stress range was between 93 and 130 MPa, and for d_{33} the range was from 132 to 178 MPa. Figures 1-5 show data for the Type I ceramics tested. The characteristics are for the first stress cycle.

The Gulton and Vernitron ceramic had the high resistance to depolarization under stress, characteristic of Type III material. However, because it was purchased as Type I, the data obtained are included in this section. One way in which Gulton and Vernitron Type I ceramics indicate their resistance to stress depolarization is in the good recovery of g_{33} and d_{33}, as compared to the other Type I ceramics, as stress is released.

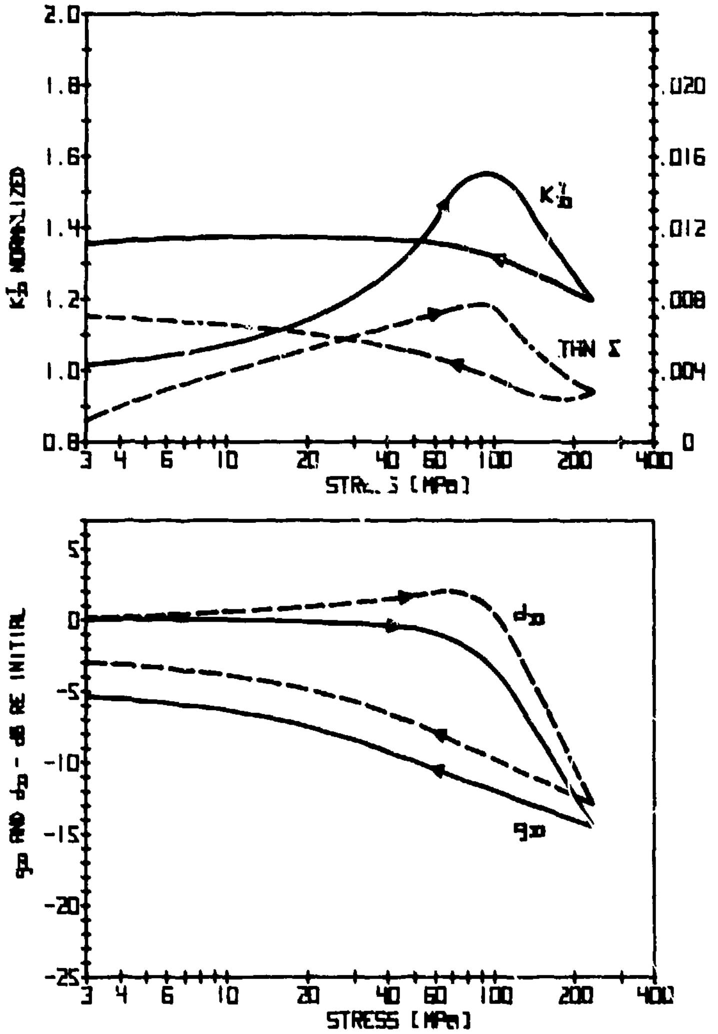

Fig. 1 — Type I ceramic (Channel 5400): dependence on stress of K_{33}^T, tan δ, g_{33}, and d_{33}

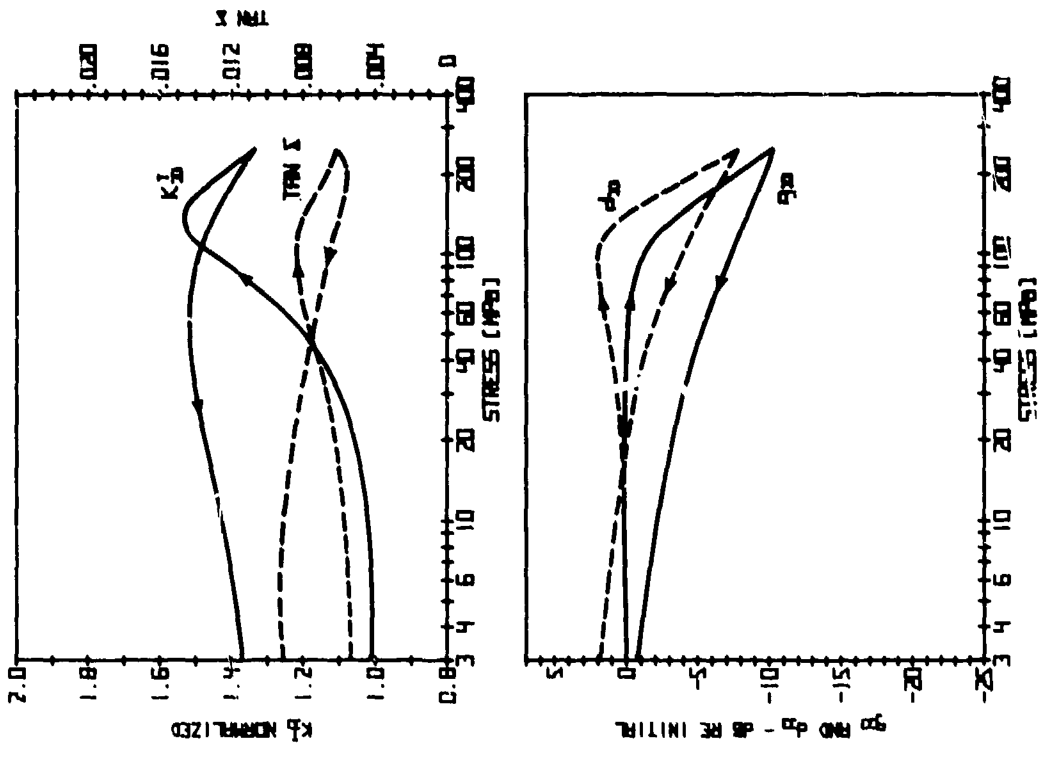

Fig. 2 — Type I ceramic (Gulton HDT-31): dependence on stress of K_{33}^T, tan δ, g_{33}, and d_{33}

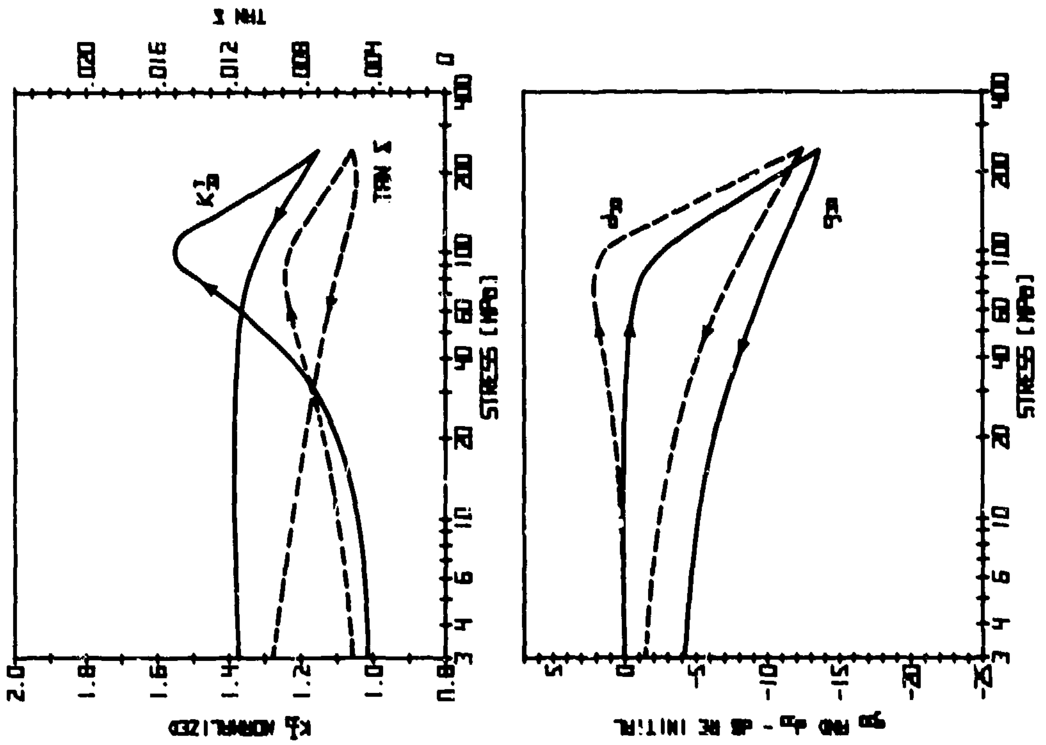

Fig. 3 — Type I ceramic (Marine Resources TCD-4): dependence on stress of K_{33}^T, tan δ, g_{33}, and d_{33}

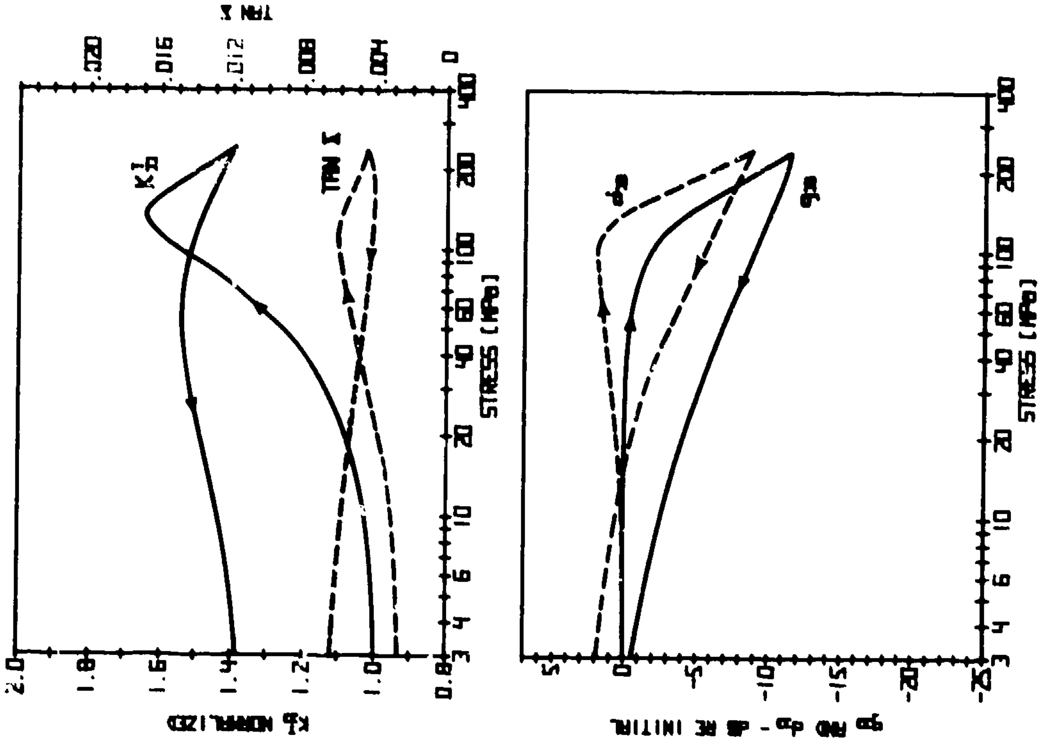

Fig. 4 — Type I ceramic (Vernitron PZT-4): dependence on stress of K^T_{33}, tan δ, g_{33}, and d_{33}

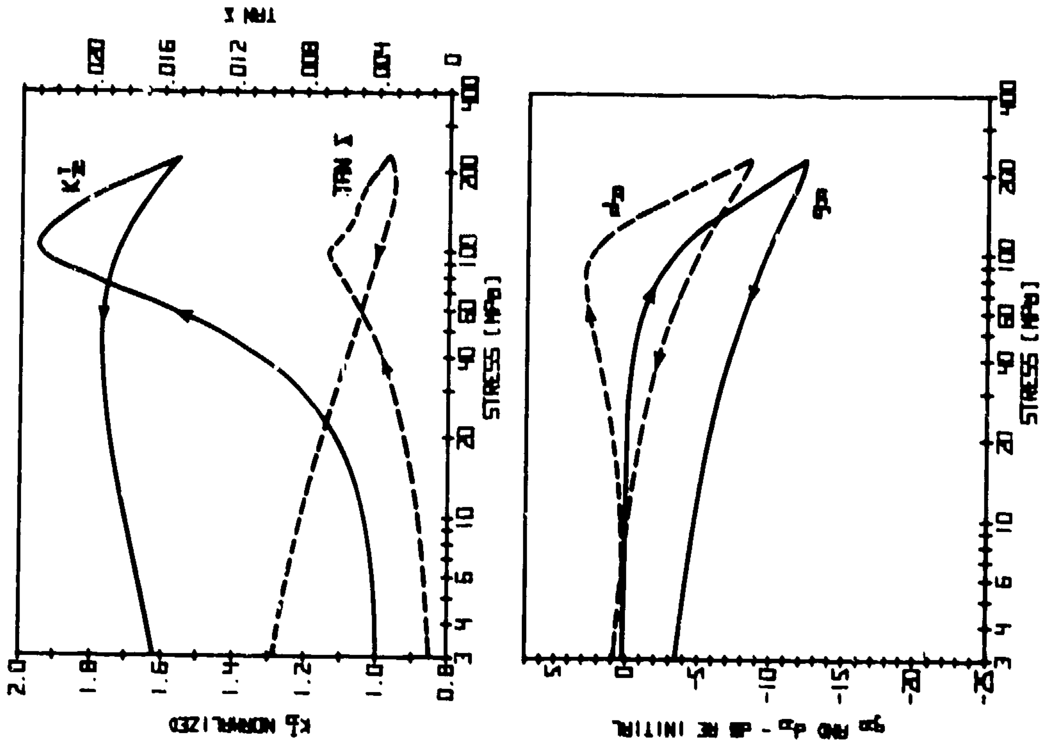

Fig. 5 — Type I ceramic (Honeywell K-12): dependence on stress of K^T_{33}, tan δ, g_{33}, and d_{33}

Type II Ceramics

Table 2 reviews the Type II materials studied. Dielectric constant K_{33}^T peaked between stresses of 52 and 62 MPa, with an average of 58 MPa. The change of K_{33}^T ranged from +17% to +31%, with an average of 21%. On most samples, tan δ consistently decreased with increasing stress. Parameter g_{33} decreased 3 dB from the initial value at stresses between 50 and 59 MPa; the corresponding points for d_{33} were between 60 and 70 MPa. Figures 6-10 show the dielectric and piezoelectric parameters of the Type II ceramics tested.

Type III Ceramics

Table 3 compares the Type III materials studied. K_{33}^T peaked at stresses between 125 and 165 MPa, with an average of 149 MPa. The percentage change of K_{33}^T ranged from +70% to +95%, with an average of 79%. Tan δ increased with increasing stress, reaching a maximum between 100 and 160 MPa; it thereafter decreased until maximum stress was attained. Stresses causing g_{33} to decrease 3 dB from initial value were 111 to 139 MPa, and for d_{33} it was 172 to 210 MPa. Figures 11-15 show the dielectric and piezoelectric parameters of the Type III ceramics tested.

COMPARISON OF MIL TYPES

Piezoelectric Constant g_{33}

Figure 16 summarizes the composite characteristics of g_{33} for military Types I, II, and III ceramics. The roll-off of this parameter with increasing stress falls within definite regions for each of the three types of ceramic. However, the region occupied by the Type I material completely overlaps the region of the Type III material. The region of g_{33} characteristics for Type I is divided into two subgroups to emphasize the ceramics having greater resistance to stress. Type II material rolls off at stress values about half that of the Types I and III material. The piezoelectric constant g_{33} decreases approximately as the negative 5/3 power of stress for all three types of ceramic in the roll-off region.

Table 2 — Comparison of Type II Ceramics

Manufacturer	Mfg. Type	Stress K_{33}^T (max) (MPa)	K_{33}^T Percentage Change Initial-To-Peak	tan δ		Stress g_{33} (-3 dB) (MPa)	Stress d_{33} (-3 dB) (MPa)
				Max.	Min.		
Channel	5500	59	+19	0.0155	0.0078	55	63
Gulton	HST-41	60	+31	0.0158	0.0071	52	66
Marine Resources	TC[illegible]-5	52	+20	0.0159	0.0096	50	60
Vernitron	PZT-5A	62	+20	0.0163	0.0094	59	70
Edo Western	EC-65	56	+17	0.0177	0.0099	50	60
Average		58	21			53	64

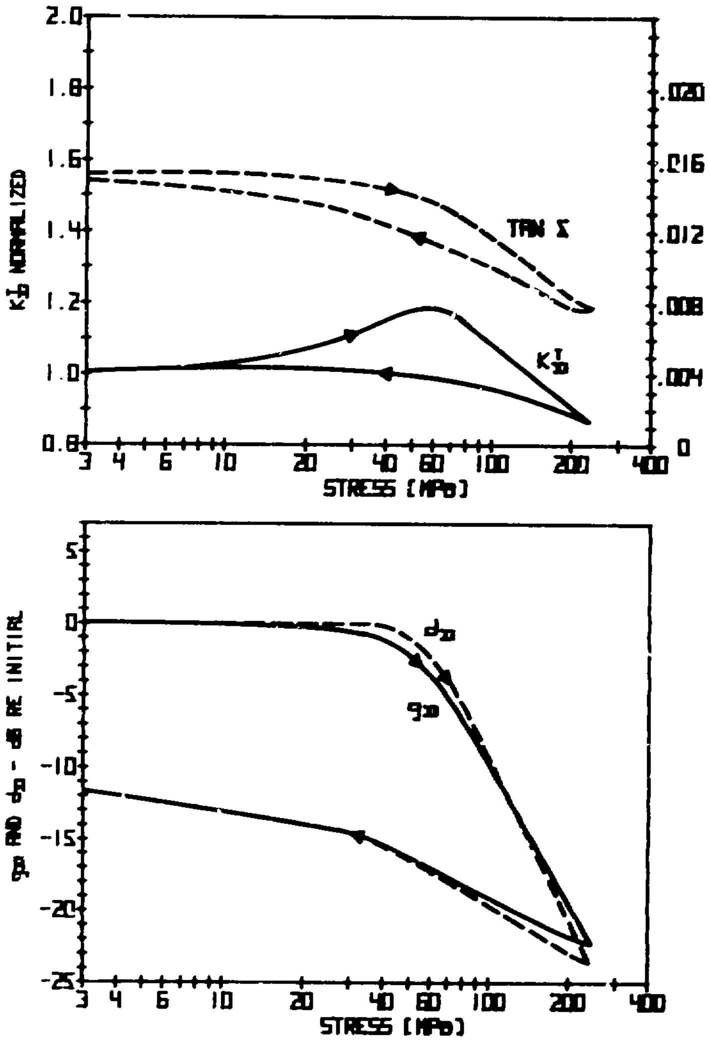

Fig. 6 — Type II ceramic (Channel 5500): dependence on stress of K_{33}^T, tan δ, g_{33}, and d_{33}

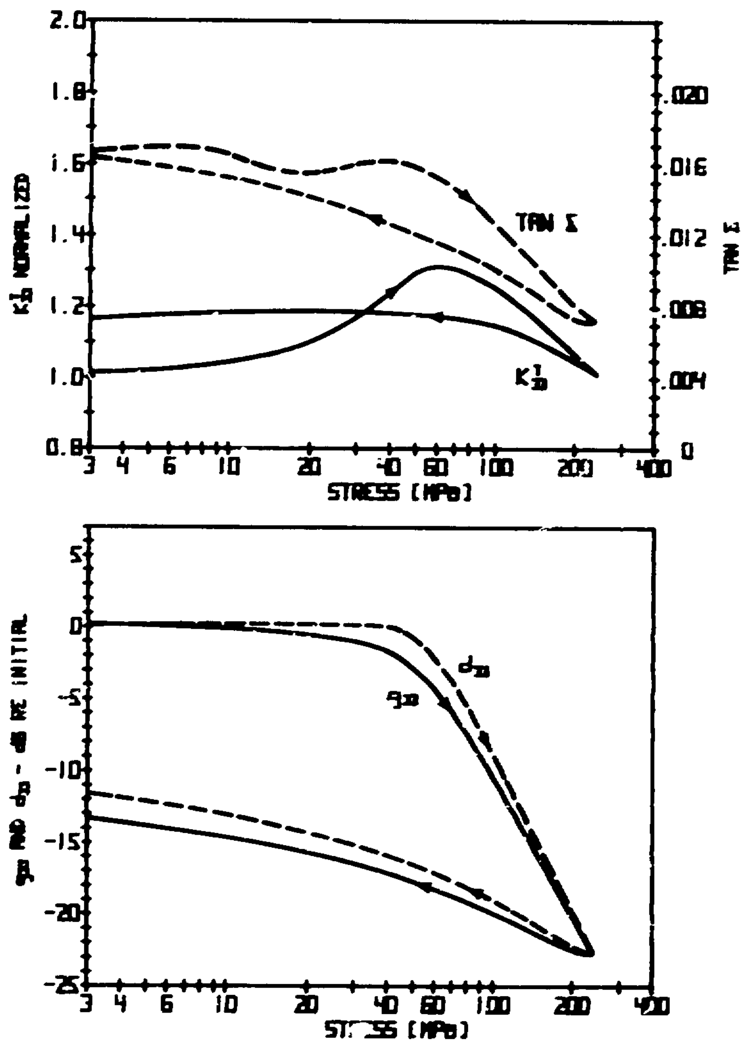

Fig. 7 — Type II ceramic (Gulton HST-41): dependence on stress of K_{33}^T, tan δ, g_{33}, and d_{33}

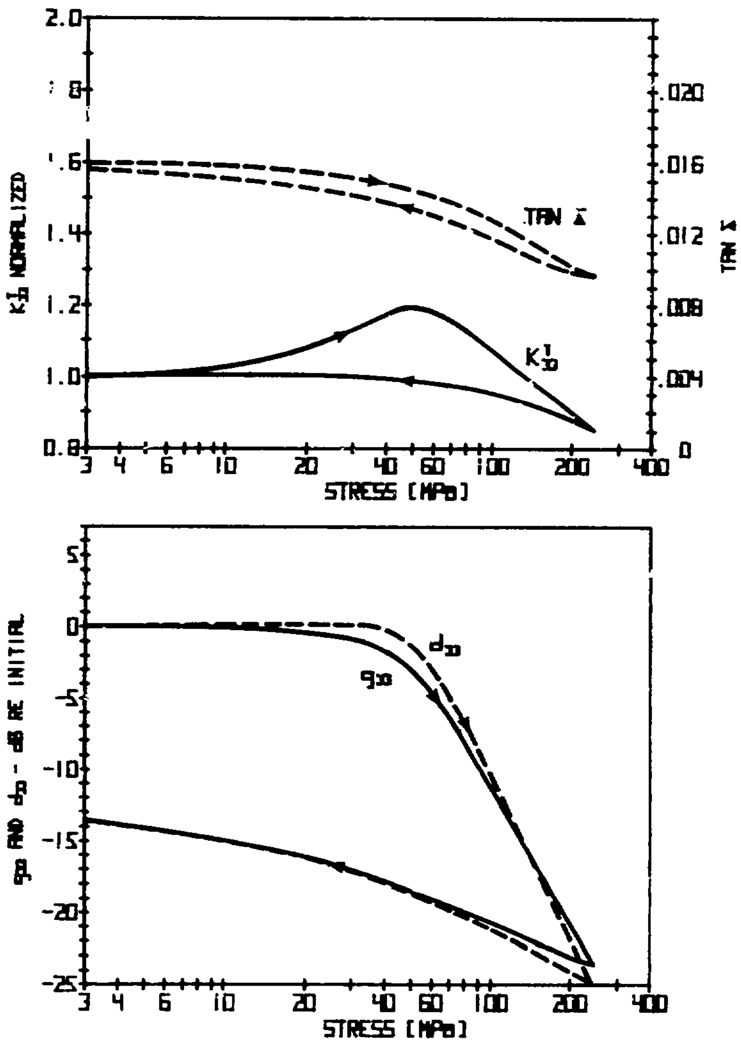

Fig. 8 — Type II ceramic (Marine Resources TCD-5): dependence on stress of K_{33}^T, tan δ, g_{33}, and d_{33}

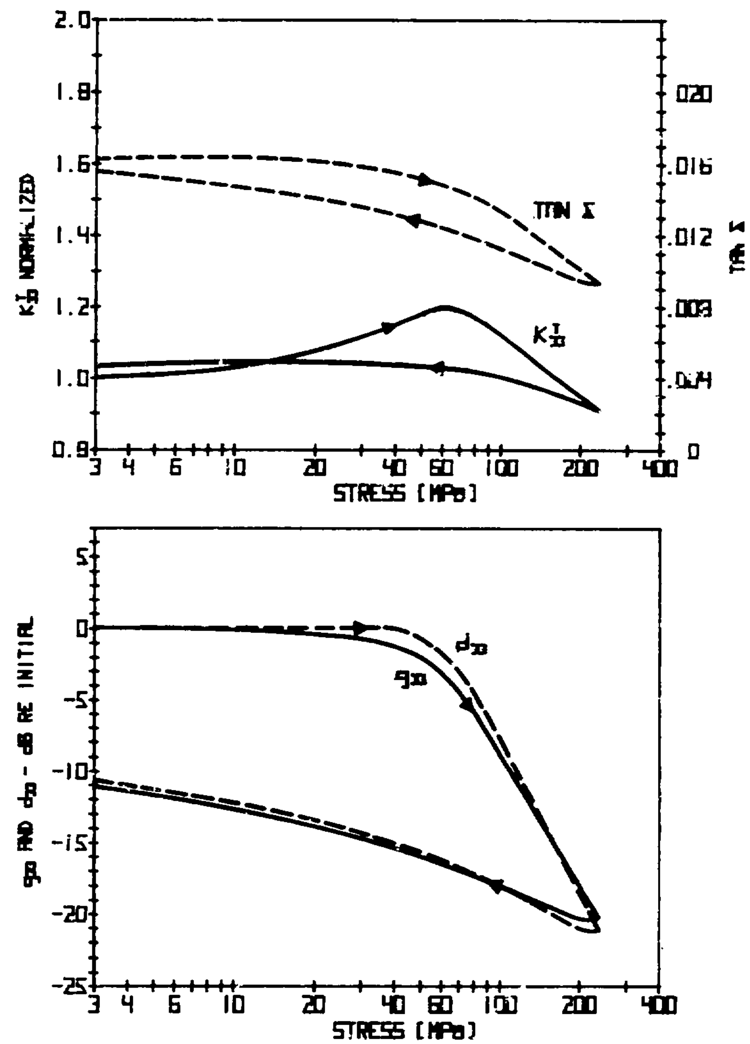

Fig. 9 — Type II ceramic (Vernitron PZT-5A): dependence on stress of K_{33}^T, tan δ, g_{33}, and d_{33}

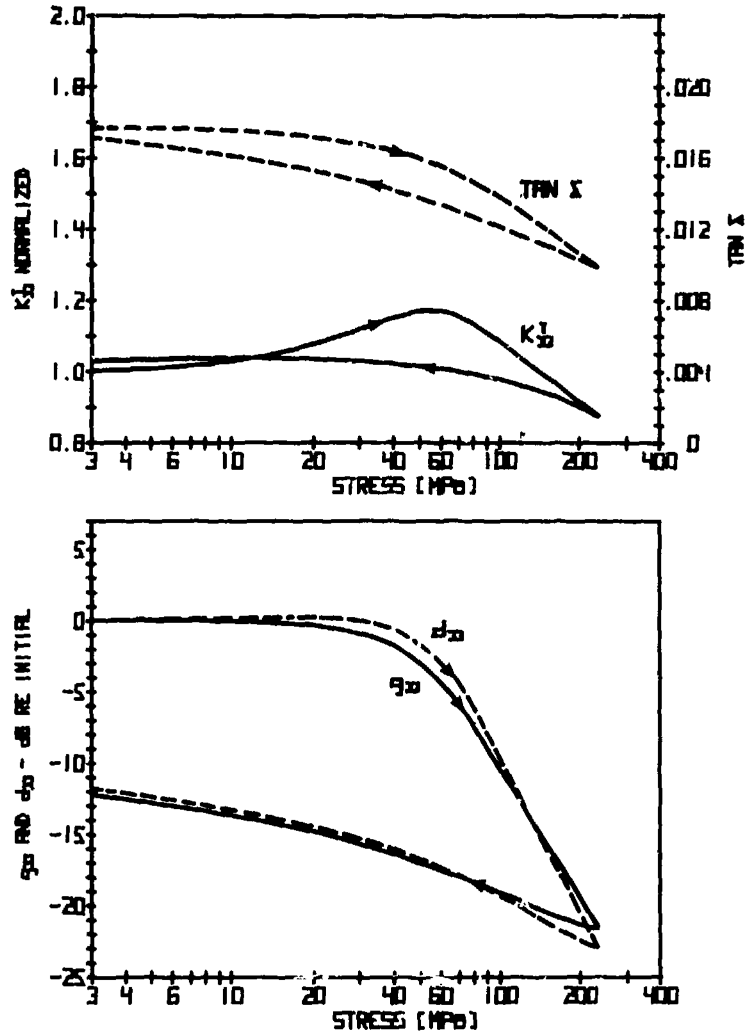

Fig. 10 — Type II ceramic (EDO Western EC-65): dependence on stress of K_{33}^T, tan δ, g_{33}, and d_{33}

Table 3 — Comparison of Type III Ceramics

Manufacturer	Mfg. Type	Stress K_{33}^T (max) (MPa)	K_{33}^T Percentage Change Initial-To-Peak	tan δ		Stress g_{33} (-3 dB) (MPa)	Stress d_{33} (-3 dB) (MPa)
				Max.	Min.		
Channel	5800	125	+95	0.0072	0.0036	115	177
Gulton	G1408	165	+70	0.0059	0.0030	139	204
Marine Resources	TCD-8	150	+75	0.0062	0.0035	111	172
Vernitron	PZT-8	162	+80	0.0064	0.0034	138	210
Edo Western	EC-69	143	+73	0.0069	0.0046	129	186
Average		149	79			126	190

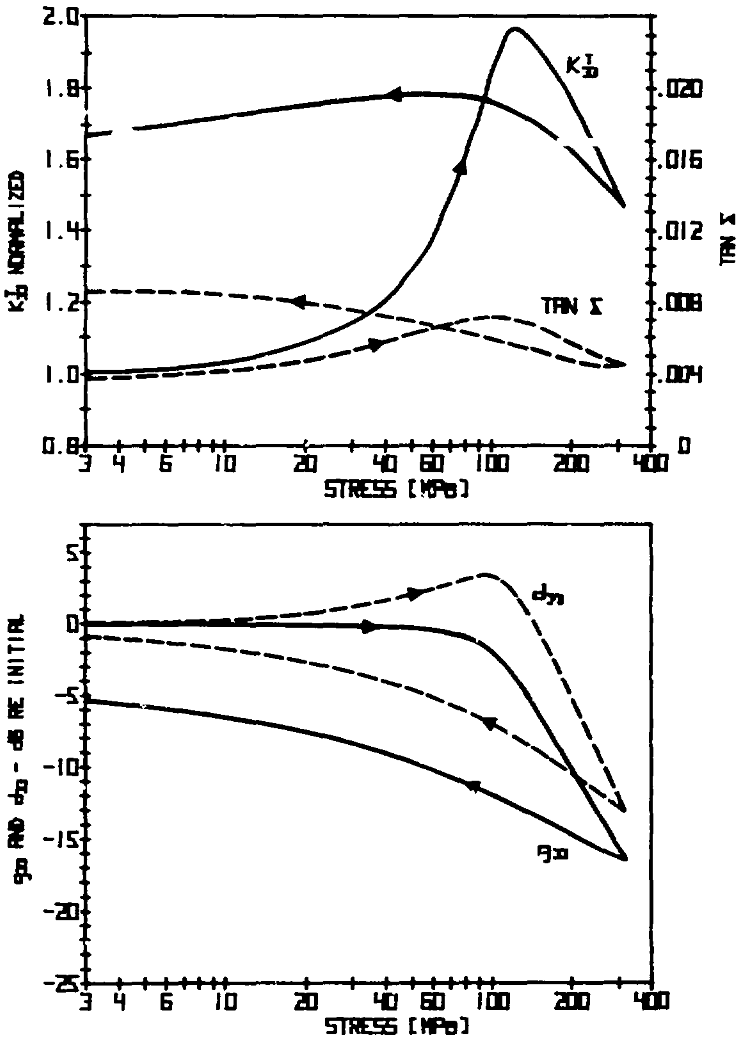

Fig. 11 — Type III ceramic (Channel 5800): dependence on stress of K_{33}^T, tan δ, g_{33}, and d_{33}

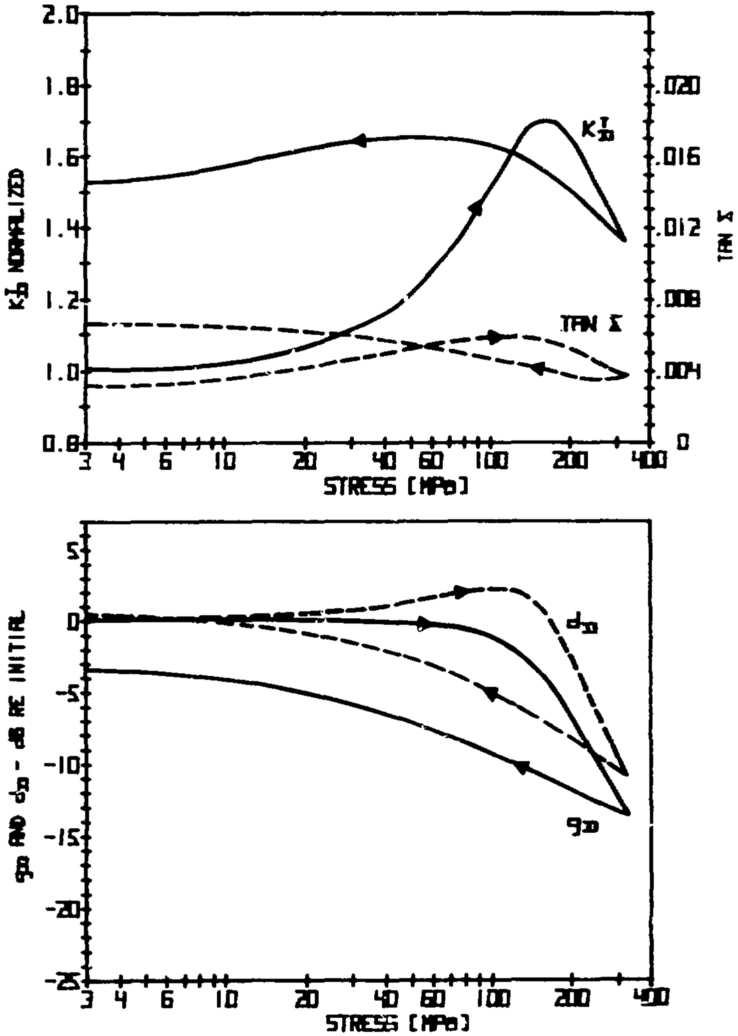

Fig. 12 — Type III ceramic (Gulton G1408): dependence on stress of K_{33}^T, tan δ, g_{33}, and d_{33}

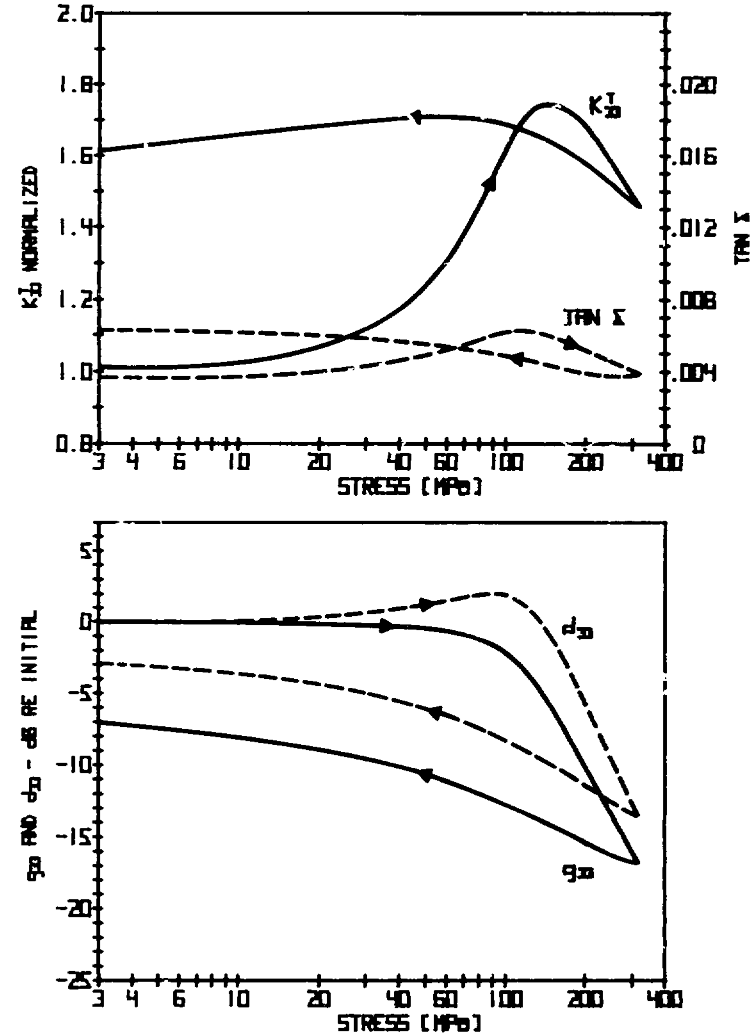

Fig. 13 — Type III ceramic (Marine Resources TCD-8): dependence on stress of K_{33}^T, tan δ, g_{33}, and d_{33}

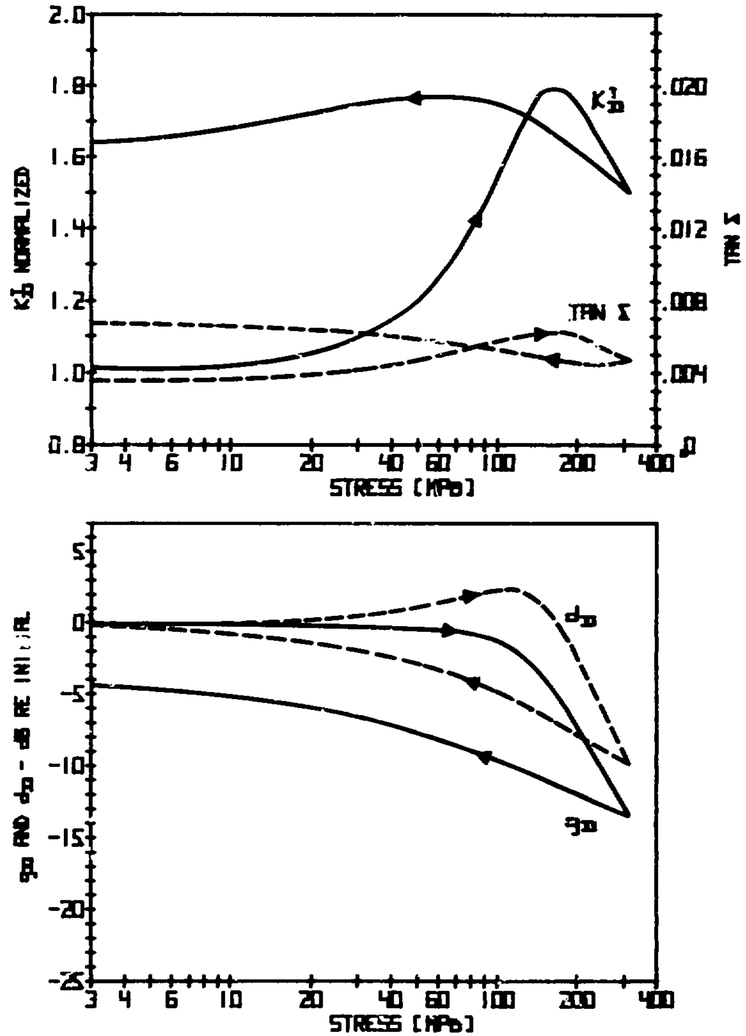

Fig. 14 — Type III ceramic (Vernitron PZT-8): dependence on stress of K_{33}^T, tan δ, g_{33}, and d_{33}

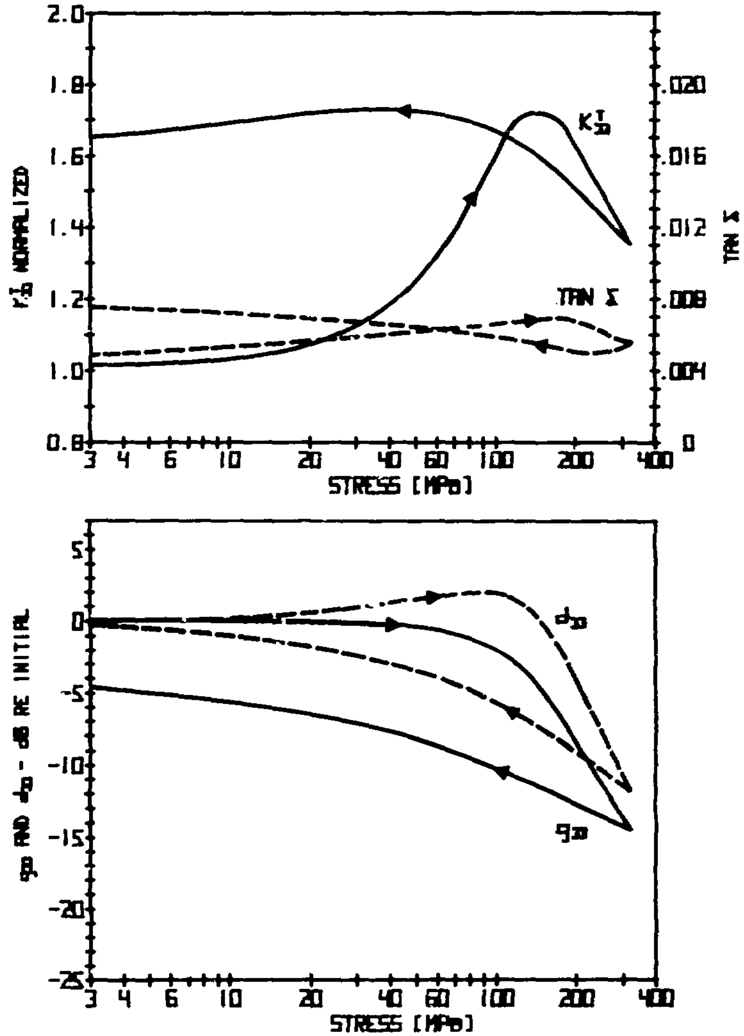

Fig. 15 — Type III ceramic (EDO Western EC-69): dependence on stress of K^T_{33}, tan δ, g_{33}, and d_{33}

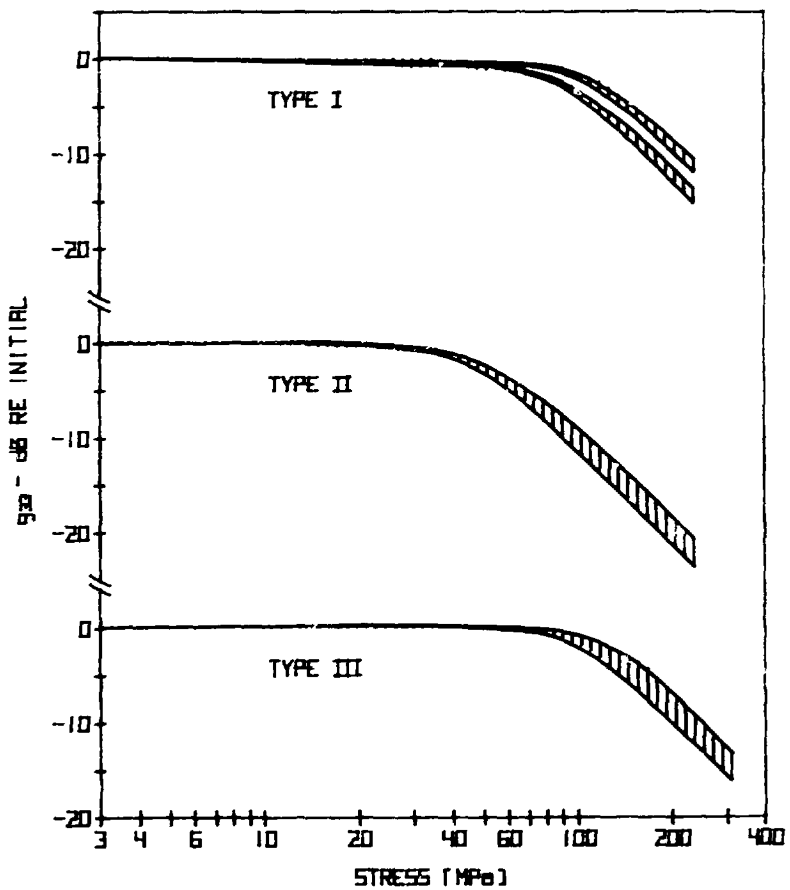

Fig. 16 — Piezoelectric constant g_{33} comparison

Dielectric Constant K_{33}^T

Figure 17 summarizes the composite characteristics K_{33}^T. There was a substantial spread of the peaks of K_{33}^T as a function of stress for each of the three types of material. However, as an average the Type II material peaked at a stress about half that of Type I material. Type III material peaked at an average stress about 30% higher than the Type I material. Type III material had the highest relative increase of K_{33}^T, followed by Types I and II decreasing successively. At stresses higher than the peak, K_{33}^T is approximately a function of the negative cube root of the stress.

Piezoelectric Constant d_{33}

The constant d_{33} is the product of g_{33} and K_{33}^T, as shown in Eq. (3). Type II material exhibited only a small maximum with stress, but Types I and III both had noticeable peaks of similar relative amplitude. There was considerable overlap in the d_{33} curves of the different samples of Types I and III. The Type III material had slightly higher maxima at slightly

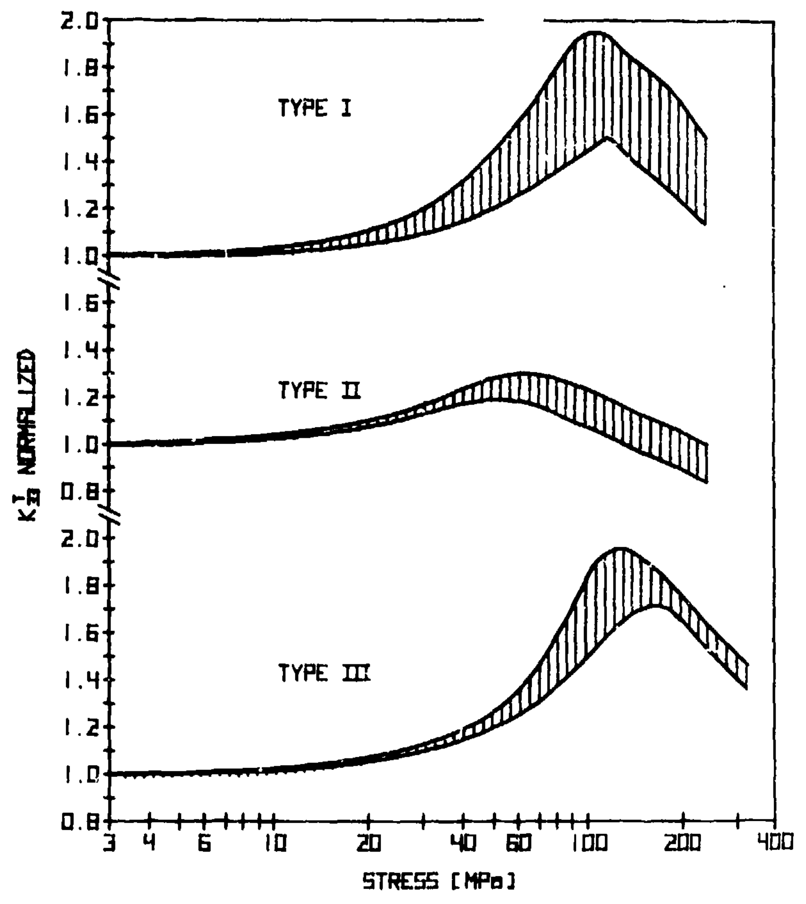

Fig. 17 — Dielectric constant K_{33}^T comparison

higher stress. The decrease of d_{33} above the peak is related approximately to the inverse square of the stress.

Dielectric Loss Tangent tan δ

Comparison of the stress effects on tan δ for the three types of material reveals similarity between Types I and III and a difference between these types and Type II. The magnitude of tan δ and the peak in the stress response of tan δ were approximately the same for Types I and III, with the changes associated with Type I materials being slightly greater. For the Type II materials tan δ showed a continual decrease with increasing stress. The decrease of tan δ for the Type II materials above 100 MPa is related to the inverse square root of stress, with variations ranging from $T^{-1/3}$ to $T^{-2/3}$.

CONCLUSION

This series of stress effect tests of the three types of lead zirconate titanate produced by several manufacturers shows the degree of variation to be expected between and within MIL types. There is considerable similarity between the stress characteristics of MIL Types I and III material, with Type III being a little more stress resistant than Type I. However, some manufacturers' Type I material is more stress resistant than Type III of other manufacturers. The shapes of parameter characteristic curves for Type II material are basically similar to those for Types I and III materials, but the stress resistance of Type II is 40% to 50% less than that of the other types.

In a hydrophone, these ceramics will probably operate reliably under one-dimensional stress up to a maximum determined by the peak of the K_{33}^T dielectric curve. Approximate maximum stress values are 100 MPa for Type I, 55 MPa for Type II, and 125 MPa for Type III. Degradation of g_{33} may result if the ceramic is stress cycled to higher stresses than these. However, selected Type III ceramic may be used safely up to 165 MPa.

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